HEAT TRANSFER IN AN INTUMESCENT MATERIAL USING A THREE-DIMENSIONAL LAGRANGIAN MODEL

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International Conference on Fire Research and Engineering, September 10-15, 1995. Orlando, FL. Proceedings. Sponsored by National Institute of Standards and Technology (NIST) and Society of Fire Protection Engineers (SFPE). D. Peter Lund and Elizabeth A. Angell, Editors. Society of Fire Protection Engineers, Boston, MA, 1995.

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Introduction

Intumescence is a property of an important class of fire-resistant materials. In the presence of fire, a succession of chemical reactions results in melting, the generation of multiple tiny bubbles causing the material to swell, and solidification into a thick multicellular char layer.[1] An intumescent coating protects the underlying substrate from fire through two mechanisms: heat is absorbed by the endothermic chemical reactions that produce the bubbles, and the low thermal conductivity of the bubbles provides an insulating layer. The chemical mechanism causes a plateau in the plot of substrate temperature vs. time, and the decreased effective thermal conductivity slows the temperature increase with time for the final char layer.

The timing of events as the material is heated is critical to its protective qualities, but the basic physical, thermal, and chemical mechanisms and their interactions are still not well understood. The design of intumescent materials has therefore traditionally proceeded through systematic testing of a variety of chemical formulations.

There have been attempts to develop a better understanding of intumescence. Previous models[2–6] have treated the intumescent system as a one-dimensional heat transfer problem through a single layer of time-varying thickness or through three layers consisting of virgin material, a thin pyrolysis zone, and char. The swelling of the material, observed to be an important factor in the calculations, is derived from experimental data and entered as an input to the models.

In this model, swelling and heat transfer are determined from fundamental principles. The goal of this research is to provide a design tool to aid in optimization of these materials.

The Intumescent Model

In the three-dimensional, time-dependent numerical model under development, the intumescent material is simulated by a volume in which many bubble sites (on the order of 10,000 in 100 cm^3) are randomly distributed. At time $t = 0$ a heat flux is applied to the upper surface. As the temperature within the volume increases, the viscosity of the material decreases and bubbles nucleate, swell, and migrate. The small Reynolds number, the dominance of swelling over migration, and the small size of the bubbles compared to their separation during early stages of swelling enable the flow field to be approximated as the sum of flow fields for individual bubbles.[7] Since the individual flow fields can be expressed analytically, a large number of bubbles can be handled on an engineering workstation.

During development of the hydrodynamic submodel, heat transfer calculations were decoupled from swelling. A simple expression for temperature as a function of depth and time was used to trigger nucleation of each bubble. A time sequence showing the results of this procedure is presented in Figure 1. In the next phase of model development, the temperature field is influenced at each bubble site by the endothermic heat of chemical reaction and the small thermal conductivity within the bubble. This talk will show the results of representing the temperature field as the summation of fields due to individual bubbles, in a similar manner to the hydrodynamic calculations.

Heat Transfer for a Single Bubble

By analogy with the approach used to determine the hydrodynamics of the intumescent material, the temperature field can also be determined as a summation of fields due to the presence of individual bubbles. The problem to be solved is a slowly growing and migrating sphere in a fluid of infinite extent. The sphere is thermally conducting, and there is a source or sink of heat on its surface due to chemical reactions. In the absence of the sphere, the background temperature varies linearly with depth.

In an intumescent material, where the Reynolds number is very small, the timescales for the swelling and migration of each bubble are much longer than the timescale for thermal diffusion. This is true for both the intumescent melt and the bubble. We can therefore assume that the problem is quasi-steady, reducing the energy equation to a simple Laplace equation with boundary conditions that take the background temperature gradient and the chemical heat source into account. This results in a simple analytical expression for the temperature field of an individual conducting sphere. The important thermal mechanisms acting to reduce the temperature at the substrate beneath the intumescent material can be seen by considering the effects of chemical reactions and thermal conductivity differences in turn.

To observe the effects of an exothermic or endothermic reaction taking place on the surface of the sphere, the thermal conductivities of sphere and background materials are set to equal values. The temperature contours for these reactions are shown in Figure 2. For the

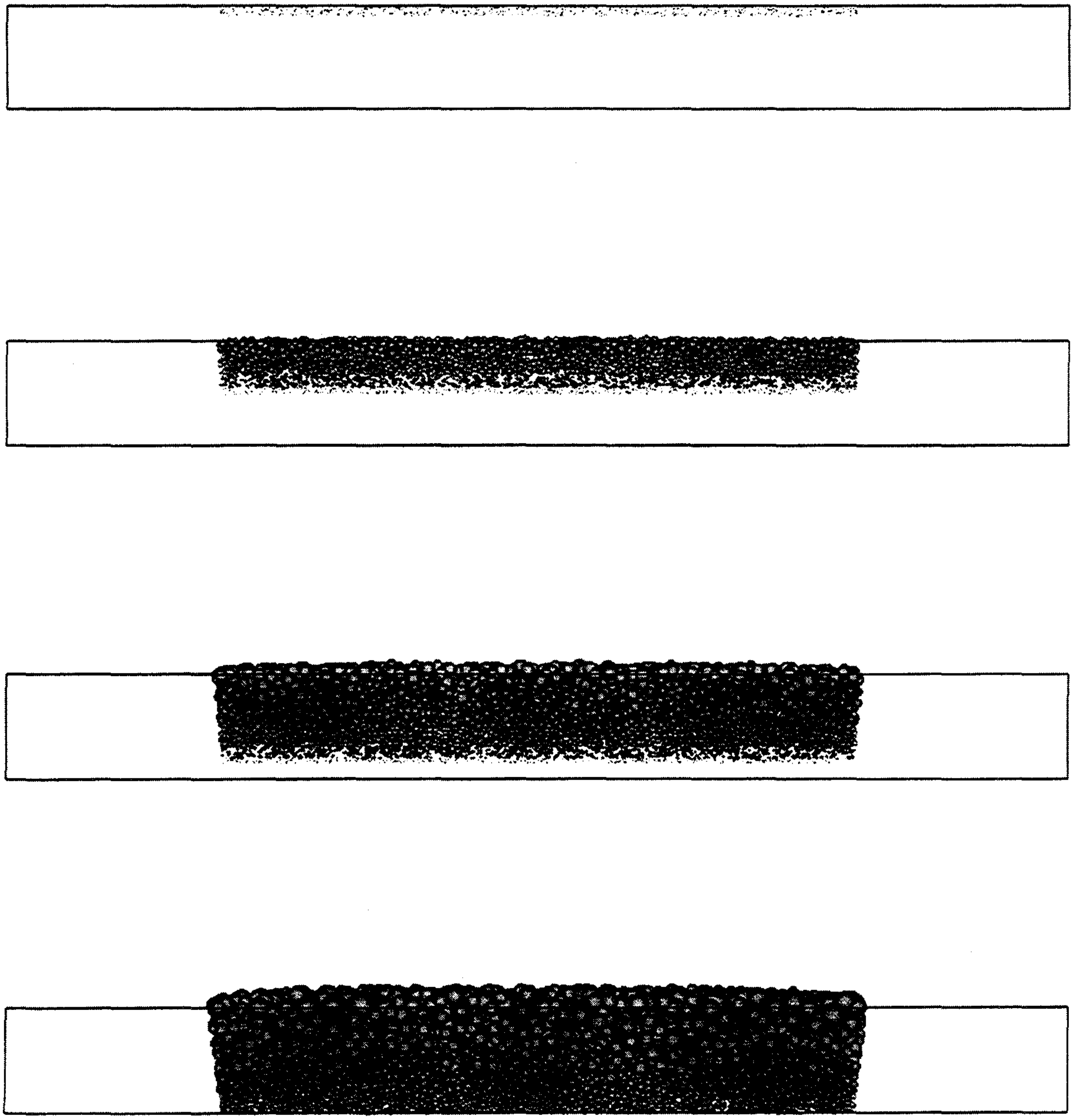

Figure 1: Development of 10,000 bubbles with time as a heat flux of 40 kW/cm^2 is applied on the upper surface. The sample is initially 10 cm $\times$ 10 cm $\times$ 1 cm thick.

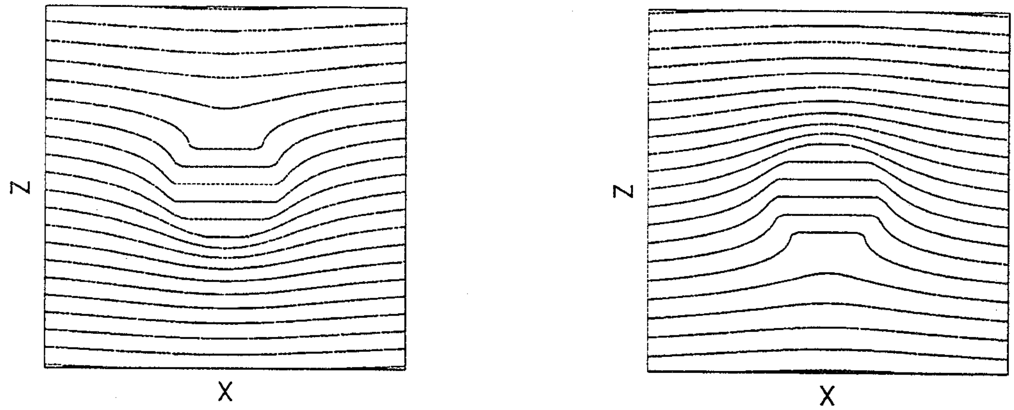

Figure 2: **Temperature contours in the X–Z plane for an exothermic reaction (left) and an endothermic reaction (right) taking place at the surface of a sphere. The sphere is located at the center of the plot. Temperature increases in the positive Z direction.**

exothermic reaction, the temperature is higher than the background temperature field both above and below the sphere, as expected. For the endothermic reaction, temperatures are lower.

The effect of the thermal conductivity of the sphere in comparison with the surrounding fluid, in the absence of a heat source, is shown in Figure 3. For a sphere with higher thermal conductivity than the surrounding fluid, such as a drop in air, the temperature is nearly uniform within the sphere. The temperature of the surroundings is increased below the drop and decreased above. For a bubble, with lower thermal conductivity than its surroundings, the temperature is decreased below the bubble and increased above.

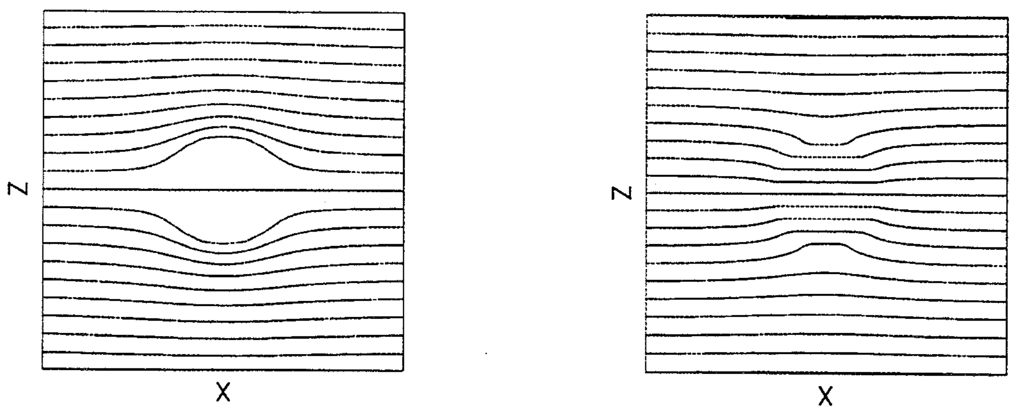

Figure 3: **Temperature contours for a drop with thermal conductivity ten times that of its surroundings (left), and a bubble with one-tenth the thermal conductivity of its surroundings (right).**

The solution of the full intumescent problem with many bubbles is aided by the fact that the temperature field external to each sphere can be represented mathematically by singularities located at the sphere center. The exterior temperature field due to chemical reactions is identical to that generated by a source or sink, and the exterior field due to differences in thermal conductivity is identical to a dipole field.

Multiple Bubbles in an Intumescent Material

The heat transfer problem that must be solved is an energy equation for the intumescent melt subject to a heat flux from the fire at the upper surface, appropriate (such as adiabatic or insulating) boundary conditions at the substrate on the lower surface, and temperature and heat flux boundary conditions at each bubble surface. For the number of bubbles ($\sim 10,000$) needed in this simulation, an exact solution of this problem is not possible. The essential physics, however, are captured by considering each bubble to be under the influence of its local temperature and temperature gradient, such that the total temperature field is represented by the sum of fields from individual bubbles with the background field. Like the hydrodynamic submodel, this approximation is best at early times when the distance between bubbles exceeds their size.

Figure 4 shows the temperature contours from the sum of three bubble fields with a linear background field. Both endothermic reaction and thermal conductivity mechanisms are acting to reduce the temperature beneath the bubbles. The figure on the right shows the field generated by the equivalent sink and dipole singularities.

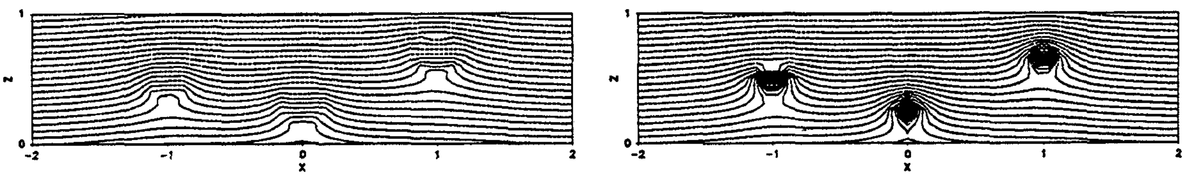

Figure 4: **Temperature contours for three bubbles with low thermal conductivity and endothermic chemical reactions taking place on each bubble surface. On the left is the analytical solution, and on the right is a summation of discretized sink and dipole singularities.**

As the bubbles nucleate and swell within the intumescent material, the model geometry becomes increasingly distorted from the initial rectangular solid, as illustrated in Figure 1. The energy equation is conveniently solved in a rectangular geometry with flat boundaries at top and bottom. This can be arranged by tracing the locations of convected gridpoints back in time to their original positions through the introduction of a Lagrangian coordinate system.[8] The Lagrangian formulation eliminates the convection term from the energy equation, the complex geometry is incorporated into the diffusion coefficient, and sink and dipole singularities added at each bubble site enable the appropriate boundary conditions to be met at each bubble. The temperature field is a finite difference solution of the energy equation with a heat flux applied at the top surface and adiabatic or other conditions applied at the lower boundary.

A comparison of substrate temperature as a function of time with the behavior of real intumescent materials will be shown in this talk.

Conclusions

Modelling intumescence as the summation of flow and temperature fields from a large number of individual bubbles shows great promise for improving our understanding of the basic physics and chemistry behind this behavior. The two basic mechanisms providing thermal protection are easily incorporated into this model through a sink singularity, which represents the endothermic chemical reactions during gasification, and a dipole singularity, representing the difference in thermal conductivity between the bubble and the intumescent melt. The time-dependent heat transfer problem for the swelling intumescent material is readily solved by using a Lagrangian coordinate system to set up a finite difference problem in the original rectangular geometry.

This model is efficient enough to be implemented on an engineering workstation.

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